

THE EFFECT OF ADSORBENT ADDITIVE ON CLAY BASED ADSORBENT TO THE REMOVAL OF MULTIDYE SYSTEM

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ABSTRACT

The use of cheap and efficient additives has been studied to find an alternative substitution of expensive additives that is used for the removal of dyes from wastewater. Powdered Activated Carbon (PAC) is one of the most expensive yet effective additives. The main reason doing this research is to find the cheapest and effective adsorbent to treat the waste water. The objective of this experiment is to study the effect of adsorbent additives on clay based adsorbent to the removal of multi-dye system. The scopes are to study the influence of pH towards dye removal performance and to study the effect of multi-dye removal by adding different ratio of additive with clay. Clay is used as the adsorbent and four additives are used to increase the efficiency of dyes removal from wastewater which is Zeolite powder, PAC, Chitosan, and Magnesium Chloride. In the study of determining the effective pH towards dye removal, the mass of clay and additives are set to constant and were added into beaker containing 250ml of dyes solution. The range of pH that used is between 1-14. The results show that Zeolite powder, PAC, and Chitosan are effective at acidic solution which is at pH 5 while Magnesium Chloride is effective at pH 11 which is in basic solution. To study the effect of multi-dye removal with different ratio of additives, the experiments are conducted by adding different amount of additive into a constant amount of clay. The mixture is then repeated on dye removal experiment. From the research, result shows that PAC is the most effective additives compared to others which have high percentage of dyes removal for Methylene Blue at 98.98% and Methyl Orange at 99.19% in acidic solution. The result shows that the best percentage of dye removal occur when the ratio of clay and additives are similar. However, PAC is still proven to be the best additives among all which show the highest percentage removal of Methylene Blue at 99.58% and Methyl Orange at 98.86% %. For the conclusion, PAC is the most effective additive used for the removal of dyes in the solution of 1.0g clay and 1.0g PAC at pH 5 which is in acidic solution. Further research should be done to look for the cheapest alternative for PAC.

ABSTRAK

Penggunaan pemangkin yang murah dan berkesan telah dikaji untuk mencari alternative bagi menggantikan pemangkin yang mahal untuk merawat air kumbahan. Serbuk karbon reaktif (PAC) merupakan salah satu pemangkin yang mempunyai harga pasaran yang tinggi tetapi sangat berkesan. Tujuan utama kajian ini dijalankan adalah untuk mencari penjerap yang murah dan berkesan untuk merawat sisa air. Objektif eksperimen ini adalah untuk mengkaji kesan pemangkin pada tanah liat yang bertindak sebagai penjerap untuk mengeluarkan warna dari sisa air. Skop kajian ini adalah untuk mempelajari pengaruh pH terhadap prestasi penghapusan pewarna dan untuk mempelajari pengaruh penghapusan multi-dye dengan menambah nisbah pemangkin yang berbeza dengan tanah liat. Tanah liat digunakan sebagai agen penjerap dan empat pemangkin digunakan untuk meningkatkan kecekapan penyisihan zat warna dari sisa cair iaitu serbuk Zeolite, PAC, Chitosan, dan Magnesium Klorida. Dalam kajian penentuan keberkesanan pH terhadap penghapusan pewarna, berat tanah liat dan aditif ditetapkan secara sekata dan ditambah ke dalam bikar berisi 250 ml larutan pewarna. pH yang digunakan adalah antara 1-14. Keputusan kajian menunjukkan bahawa serbuk Zeolite, PAC, dan Chitosan berkesan pada keadaan asid iaitu pada pH 5, sedangkan Magnesium Klorida adalah berkesan pada pH 11 yang dalam larutan alkali. Untuk mempelajari pengaruh penghapusan multi pewarna dengan nisbah yang berbeza dari pemangkin, percubaan dilakukan dengan menambah perbezaan jumlah pemangkin ke dalam jumlah yang konstan dari tanah liat. Campuran ini kemudian diulang. Dari hasil kajian menunjukkan hasil bahawa PAC merupakan pemangkin yang paling berkesan berbanding dengan orang lain yang mempunyai peratusan tinggi pengeluaran pewarna untuk Metilen Blue di 98.98% dan Metil Orange sebanyak 99.19% dalam larutan asid. Keputusan kajian menunjukkan bahawa peratusan terbaik pengeluaran pewarna terjadi ketika nisbah dari tanah liat dan aditif yang serupa. Namun, PAC masih terbukti menjadi aditif terbaik di antara semua yang menunjukkan penghapusan peratusan tertinggi Metilen Blue di 99.58% dan Metil Orange di 98.86%. Kesimpulannya, PAC adalah yang pemangkin paling berkesan digunakan untuk menghilangkan zat warna dalam larutan tanah liat pada nisbah 1.0g:1.0g pada pH 5 yang dalam larutan asid. Penelitian lebih lanjut perlu dilakukan untuk mencari alternative termurah selain PAC.

TABLE OF CONTENTS

CHAPTER	TITLE	PAGE
	TITLE	i
	STUDENT DECLARATION	ii
	DEDICATION	iii
	ACKNOWLEDGEMENT	iv
	ABSTRACT	v
	ABSTRAK	vi
	TABLE OF CONTENT	vii
	LIST OF TABLES	x
	LIST OF FIGURES	xii
	LIST OF SYMBOLS/ABBREVIATION	xiii
	LIST OF APPENDICES	xv

1. INTRODUCTION

1.1	Research Background	1
1.2	Objective	7
1.3	Problem Statement	7
1.4	Scopes Of Studies	9
1.5	Rationale	9

2. LITERATURE REVIEW

2.1	Wastewaters From Industries	11
2.2	Adsorption Process	13
2.2.1	Adsorbent	16
2.2.2	Factors Affecting Adsorption	16
2.3	Clay As Adsorbent	17
2.3.1	Clay Minerals	20
2.3.2	The Crystal Structure of Clay Minerals	21
2.3.3	Crystal	22
2.3.4	Crystal Shape	22
2.3.5	Crystal Size	23
2.4	Additives Used	23
2.4.1	Zeolite	24
2.4.2	Activated Carbon	25
2.4.3	Chitosan	26
2.4.4	Magnesium Chloride	28
2.5	Dyes	29
2.5.1	Types of Dyes Used	
2.5.1.1	Methylene Blue	33
2.5.1.2	Methyl Orange	34
2.6	Analysis Of Residual	
2.6.1	Batch Analysis	35
2.6.2	UV-Vis Spectrophotometry	36

3. METHODOLOGY

3.1	Preparation of Adsorbent-Clay	38
3.2	Adsorption Process	
3.2.1	To Study The Effect of pH On Wastewater	39
3.2.2	To Study The Effect Of Dosage Of Additives	40

4. RESULT & DISCUSSION	41
4.1 Standard Curve for Stock Solution	
4.2 The Effect of pH Towards Dye Removal	
4.2.1 Percentage Removal Of Methylene Blue And Methyl Orange	
4.2.2 Percentage Removal Of Dyes For Each Additive for the effect of pH	
4.3 The effect of dosage towards dye removal	
4.3.1 Percentage Removal Of Methylene Blue And Methyl Orange	
4.3.2 Percentage Removal Of Dyes For Each Additive for the effect of dosage	
4.4 Langmuir And Freundlich Isotherm	
4.4.1 Effect of pH on dyes removal	
4.4.1.1 Table of Langmuir and Freundlich Equation	
4.4.2 The Effect Additives Dosage towards dye removal	
4.4.2.1 Table of Langmuir and Freundlich Equation	
5. CONCLUSION & RECOMMENDATION	67
6. REFERENCES	69
7. APPENDICES	71

LIST OF TABLES

TABLE NO.	TITLE	PAGE
1.1	Industrial water consumption	5
2.1	Differences between chemisorptions and physisorption	15
2.2	The Specification Of Chitosan	27
4.1	Table of standard curve for stock solution	41
4.2	Table of percentage removal of methylene blue and methyl orange for PAC	42
4.3	Table of percentage removal of dyes for PAC	43
4.4	Table of percentage removal of dyes for Zeolite	44
4.5	Table of percentage removal of dyes for Chitosan	44
4.6	Table of percentage removal of dyes for MgCl ₂	45
4.7	Table of percentage removal of methylene blue and methyl orange for PAC	46
4.8	Table of percentage removal of dyes for PAC	48
4.9	Table of percentage removal of dyes for Zeolite	49
4.10	Table of percentage removal of dyes for Chitosan	50
4.11	Table of percentage removal of dyes for MgCl ₂	51
4.12	Table Of Additives at pH 1 For Langmuir And Freundlich	56
4.13	Table Of Additives at pH 3 For Langmuir And Freundlich	56
4.14	Table Of Additives at pH 5 For Langmuir And Freundlich	56
4.15	Table Of Additives at pH 9 For Langmuir And Freundlich	57
4.16	Table Of Additives at pH 11 For Langmuir And Freundlich	57
4.17	Table Of Additives at pH 13 For Langmuir And Freundlich	57

4.18	Table Of Langmuir And Freundlich data	58
4.19	Table Of Additives at dosage 1;0 For Langmuir And Freundlich in acidic	60
4.20	Table Of Additives at dosage 0;1 For Langmuir And Freundlich in acidic	60
4.21	Table Of Additives at dosage 1;1 For Langmuir And Freundlich in acidic	61
4.22	Table Of Additives at dosage 1;1.5 For Langmuir And Freundlich in acidic	61
4.23	Table Of Additives at dosage 1;0 For Langmuir And Freundlich in basic	61
4.24	Table Of Additives at dosage 0;1 For Langmuir And Freundlich in basic	62
4.25	Table Of Additives at dosage 1;1 For Langmuir And Freundlich in basic	62
4.26	Table Of Additives at dosage 1;1.5 For Langmuir And Freundlich in basic	62
4.27	Table Of Langmuir And Freundlich data in acidic	63
4.28	Table Of Langmuir And Freundlich data in basic	65

LIST OF FIGURES

FIGURE NO.	TITLE	PAGE
2.1	Clay	19
2.2	Zeolite and Powdered Activated Carbon (PAC)	23
2.3	Chitosan and Magnesium Chloride	23
4.1	Graph of concentration vs absorbent for standard curve	41
4.2	Graph of percentage dye removal vs pH for PAC	42
4.3	Graph of percentage dye removal vs pH for PAC	45
4.4	Graph of percentage dye removal vs dosage for PAC	46
4.5	Graph of percentage dye removal vs ratio for PAC	48
4.6	Graph of percentage dye removal vs ratio for Zeolite	49
4.7	Graph of percentage dye removal vs ratio for Chitosan	50
4.8	Graph of percentage dye removal vs ratio for MgCl ₂	51

LIST OF SYMBOLS/ABBREVIATION

SYMBOL	DEFINITION
COD	Chemical Oxygen Demand
BOD	Biological Oxygen Demand
VOC	volatile organic compounds
m	meter
cm	centimeter
mm	milimeter
MW	Mega Watt
μm	micrometer
TEM	Transmission Electron Microscope
ppm	part per million
g	gram
kg	kilogram
MgCl_2	Magnesium chloride
Ca(OH)_2	calcium hydroxide
NaCl	Sodium Chloride
Na_2SO_4	Sodium Sulphate
$^{\circ}\text{C}$	degree celcius
ISO	International Standards Organization
PAC	powdered activated carbon

UV-Vis	Ultraviolet-visible
NIR	near-infrared
L	path length
Rpm	rotary per minute
Abs	absorbent
L	litre
Q_e	adsorption density at equilibrium solute concentration c_e
C_e	concentration of adsorbate
Q_{max}	maximum adsorption capacity
K_L	Langmuir constant
K_f and n	empirical constant
R^2	correlation coefficient

LIST OF APPENDICES

APPENDIX	TITLE	PAGE
A	preparation of samples	
B	effect of dosage	
C	determine the effect of ph towards the removal of dyes	
	determine the effect of dosage towards the removal of dyes	

CHAPTER 1

INTRODUCTION

1.1 Research Background

Many industries in Malaysia such as textile, paper, high-technology, paint, pharmaceuticals, food, leather, cosmetics, tannery, printing and plastics, use various dyes in order to colour their product like batik and also consume substantial volumes of water. Among various industries, textiles industry ranks first in usage of dyes for coloration of fibre. As a result, they generate a considerable amount of wastewater colour. The effectiveness of the adsorption for dye removal from wastewater has made it an ideal alternative to other expensive treatment methods (S. A. Saad, 2007).

The textile industry is one of the most chemically intensive industries on earth, and the No. 1 polluter of clean water (after agriculture). Many textile manufacturers use dyes that release aromatic amines (e.g., benzidine, toluidine). Dye bath effluents may contain heavy metals, ammonia, alkali salts, toxic solids and large amounts of pigments that many of them are toxic. About 40 percent of globally used colorants contain organically bound chlorine, a known carcinogen. Natural dyes are rarely low-impact, depending on the specific dye and mordant used. Mordants (the substance used to "fix" the colour onto the fabric) such as chromium are very toxic and high impact. The large quantities of natural dyestuffs required for dyeing, typically equal to or double that of the fiber's own weight, make natural dyes prepared from wild plants and lichens very high impact.

Traditionally produced fabrics contain residuals of chemicals used during their manufacture which are chemicals that evaporate into the air we breathe or are absorbed through our skin. Some of the chemicals are carcinogenic or may cause harm to children even before birth, while others may trigger allergic reactions in some people. According to a June 5, 2005 article in Business Week, the population that is allergic to chemicals will grow to 60 percent by the year 2020.

Today more than 9000 types of dyes have been incorporated in the colour index. Due to their low biodegradability, a conventional biological treatment process is not very effective in treating dye wastewaters, especially the reactive dyes. Physical or chemical processes have been usually used to treat them. However these processes are costly and cannot be used effectively to treat the wide range of dye wastewater (Arvanitoyannis, 1989).

As we know, textile industry has a big pollution problem. The World Bank estimates that 17 to 20 percent of industrial water pollution comes from textile dyeing and treatment. They have also identified 72 toxic chemicals in water solely from textile dyeing, 30 of which are cannot be removed. This represents an appalling environmental problem for the clothing designers and other textile manufacturers. With consumers eager to purchase eco-friendly products, water pollution from dye houses and coloration treatments could be a major hurdle for apparel manufacturers. Some companies have taken action and removed dyes from certain garments, but it seems unlikely that everyone would be happy with off-white or beige as the only choices at the store. Consumers want colour and variety in their clothing. When creating eco-friendly clothing, drapes, or even carpet, it is important not to forget the role dye plays as an environmental ill. Consumers are becoming quite conscious of how bad traditional textile dyeing is for the environment but have put up with it until now because there has not been a viable alternative.

Textile industries produce huge amounts of polluted effluents that are normally discharged to surface water bodies and groundwater aquifers. These wastewaters cause damages to the ecological system of the receiving surface water and create a lot of disturbance to the groundwater resources. Most dyes used in

textile industries are stable to light and are not biodegradable. In order to reduce the risk of environmental pollution from such wastes, it is necessary to treat them before discharging to the receiving environments. Considerable efforts have been made by many researchers to find appropriate treatment systems in order to remove pollutants and impurities of wastewaters emanated from different industries, in particular, textile industry (Arami, 2005).

Wastewater is any water that has been adversely affected in quality by anthropogenic influence. It comprises liquid waste discharged by domestic residences, commercial properties, industry, and/or agriculture and can encompass a wide range of potential contaminants and concentrations. In the most common usage, it refers to the municipal wastewater that contains a broad spectrum of contaminants resulting from the mixing of wastewaters from different sources.

Because of the low biodegradability of many textile chemicals and dyes, biological treatment is not always effective for textile industry wastewater. Therefore, especially for colour removal, various adsorbents and chemicals have been directly added into the activated sludge systems in certain studies (T. Enis, 2001). In order to remove hazardous materials, like dyes, adsorption is a method which has gained considerable attention in the recent past. Adsorption is such a useful and simple technique, which allows gathering of both kinetic and equilibrium data without needing any sophisticated instrument. Although many experimental works have been conducted to assess the capability and the performance of various adsorbents especially for the removal of dyes from the textile industry, little research has been done to model dye-removal process from the textile wastewaters and to evaluate the significance of the effect of major parameters on the percent of dye adsorption. Activated carbon use is limited due to its high cost (S. A. Saad, 2007). This is lead to the search of other cheaper additives such as Zeolite, Chitosan, and Magnesium Chloride.

1.1.1 Industrial Wastewater

Industrial wastewater treatment covers the mechanisms and processes used to treat waters that have been contaminated in some way by anthropogenic industrial or commercial activities prior to its release into the environment or its re-use. Most industries produce some wet waste although recent trends in the developed world have been to minimise such production or recycle such waste within the production process. However, many industries remain dependent on processes that produce wastewaters.

In certain industries some of the water used in manufacturing is exported as product such as brewing, soft drinks and liquid pharmaceuticals are obvious examples, but textile, paper, and sugar also contain significant levels of residual water. In some instances water is lost as steam from heating systems and from cooling towers. But in most cases the greater proportion of the water used by industry is discharged as effluent either to sewer or to the environment.

Industrial wastewaters, unlike domestic wastewater, are highly variable in quality depending on the nature of the manufacturing processes in use. Many have high Chemical Oxygen Demand (COD) concentrations and many of these like textile industry, food industry and papermaking wastes are of animal or vegetable origin. These have a high BOD: COD ratio indicating that they are readily biodegradable and can often be treated by the same processes as are used for domestic sewage in sewage treatment works. However, when such an industry is set up, the existing sewerage and sewage treatment infrastructure is unlikely to be capable of meeting the large increase in Biological Oxygen Demand (BOD) load and the sewage treatment works may be overloaded.

On the other hand there are many industrial wastes that contain “hard COD” by which we mean organic chemicals which are not biodegradable. Typically the BOD: COD ratio will be greater than 3. The pharmaceutical industries, plastics industries and within specific process in the textile industries produce wastewaters

of this type. Many of these wastewaters are toxic to activated sludge bacteria, making them unsuitable for treatment in domestic sewage treatment works, and are likely to be damaging to the environment. Volatile solvents may also pose explosion risks in sewers and pumping stations.

It is usually central or local government that is responsible for the protection of the environment and the introduction of legislation to prevent contamination of surface water by industrial discharges has significantly reduced.

1.1.2 Industrial Water Consumption

There is hardly any industry that does not use large volumes of water. Some typical “benchmark” water consumptions for industrial processes are given in Table 1.0 below.

Table 1.1: Industrial water consumption

Product	Unit	Consumption
coal	m ³ /te product	0.25
bread	m ³ /te product	1.3
meat products	m ³ /te product	16
milk bottling	m ³ /m ³ product	3
brewing	m ³ /te product	5
soft drinks	m ³ /te product	7
chemicals	m ³ /te product	5
steel rolling	m ³ /te product	1.9

iron casting	m ³ /te product	4
aluminium casting	m ³ /te product	8.5
electroplating	m ³ /te product	15.3
tanning	m ³ /te hide processed	60
soap	m ³ /te product	2
sugar refining	m ³ /te product	4
textile dyeing	m³ /te fabric processed	40-300
concrete	m ³ /te product	0.2
paper	m ³ /te product	54
power generation	m ³ /MWh steam	3
	cooling	60
automobile manufacture	m ³ /vehicle	5
dairy farming	litre/head/day	150
pig farming	litre/head/day	15
poultry farming	litre/head/day	0.3
schools	litre/head/day	75
hospitals	litre/head/day	175
hotels	litre/head/day	750
shops	litre/head/day	135
offices	litre/head/day	60

The values given above are very approximate and will vary with the process used, but they do serve to indicate just how much water industry uses. They also show that many industries which are growing in developing countries which is

particularly those associated with food processing, tanning and textiles which are those which use most water. These industries place most stress on what is likely to be a barely adequate water supply infrastructure.

1.2 Objective

The objective of this research is to study the effect of adsorbent additives on clay based adsorbent to the removal of multi dye system.

1.3 Problem Statement

At present, large amounts of highly coloured wastewater are discharged from textile, printing, paper, and leather industries which use many kinds of artificial composite dyes. These dye-containing industrial wastewaters discharged into streams and river constitutes one of the major sources of water pollution. Colour waters are objectionable on aesthetic grounds for drinking and other agricultural purposes. Colour affects the nature of the water, inhibits sunlight penetration into the stream, and reduces the photosynthetic action. In addition, some dyes are either toxic or mutagenic and carcinogenic. So, these wastes must be treated prior to discharge in order to comply with the environmental protection laws for the receiving waters (Qi Wang, 2009).

The industry has been faced with the increasing fresh and wastewater charges as well as ever more stringent effluent regulations. Traditional wastewater treatment technologies have proven to be markedly ineffective for handling wastewater of synthetic textile dyes because of the chemical stability of these pollutants. Dye is the most 'difficult to treat' constituent of the textile wastewater. Reactive dyes are hardly biodegraded in an aerobic environment. Hence, their presence in wastewater is undesirable, and it is essential to remove colouring

material from effluents before being discharged in the environment (B. E. Ahmed, 2008).

Wastewater which is from dyeing industries creates a great problem of pollution because wastes are discharged into streams and cause water pollution. Most of the adsorbent and additives that provided in the market has high quality and expensive. Small industries such as textiles industries are incapable to get the adsorbent and additives because of the high price.

As synthetic dyes in wastewater cannot be efficiently decolorized by traditional methods, the adsorption of synthetic dyes on inexpensive and efficient solid supports was considered as a simple and economical method for their removal from water and wastewater. The adsorption characteristics of a wide variety of inorganic and organic supports have been measured and their capacity to remove synthetic dyes has been evaluated.

The treatment of textile effluents is of interest due to their toxic and esthetical impacts on receiving waters. While much research has been performed to develop effective treatment technologies for wastewaters containing azo dyes, no single solution has been satisfactory for remediating the broad diversity of textile wastes. The nature of waste from the textile industry depends on the type of factory, the processes being operated and the fibres used. In general, however, textile wastewater is highly coloured. Around 10-15% of all the dyes used in the industry are released into the environment during manufacture or usage.

Human and ecological health concerns have prompted the government to require textile effluent discharges to have increasingly lower colour and nitrogen levels. Despite being aware of the problem, many textile manufactures have failed to adequately remove azo dye compounds from their wastewaters.

The amount of waste produced from textile dyeing can be minimised by a number of measures, including:

- Low liquor dyeing.
- Improved dye fixation.
- Discontinuation of overflow rinsing, where possible.
- Segregation of hot/ cold effluent and heat recovery.
- Replacement of certain toxic dyestuffs.
- Banning of chlorine based carriers/ levellers.
- Reuse of dye liquors on repeat shades.
- Replacement of wasteful two stages dyeing of blends with single stage dyeing.
- Automatic dispensing and control systems.

1.4 Scope of Study

There are some important tasks to be carried out in order to achieve the objective of this study. The important scopes have been identified for this research in achieving the objective:

- To study the effect of multi dye removal by adding different ratio of additive with clay
- To study the influence of pH towards dye removal performance

1.5 Rationale

The first rationale doing this research is because of the economy. As we know, most of the additives in the market have high value and small industries are

incapable to buy it to treat their waste. So, this research is one of the initiatives to find the lower cost adsorbent and additives that are suitable and efficient to treat the wastewater.

This research also done to save the environment from the pollutions which is water pollutions that will affects the nature of the water. When the water is polluted, it will inhibit sunlight penetrations into the stream. This problem will affect the aquatic lives such as algae, fish and others because it reduces the photosynthetic and respiratory process. Water pollution are also toxic because it change the colour, odour and taste of water that can cause harmful to human life such as allergies, skin irritations and others.

Chapter 2

Literature Review

2.1 Wastewater from Industries

The textile industry is a significant contributor to many national economies, encompassing both small and large-scale operations worldwide. In terms of its output or production and employment, the textile industry is one of the largest industries in the world. The textile manufacturing process is characterized by the high consumption of resources like water, fuel and a variety of chemicals in a long process sequence that generates a significant amount of waste. The common practices of low process efficiency result in substantial wastage of resources and a severe damage to the environment. The main environmental problems associated with textile industry are typically those associated with water body pollution caused by the discharge of untreated effluents. Other environmental issues of equal importance are air emission, notably Volatile Organic Compounds (VOC)'s and excessive noise or odour as well as workspace safety.

Textile industry is one of the leading consumers of water. It consumes about 3.2% of total consumption of water for various processes such as sizing, scouring, bleaching, dyeing, printing and other finishing processes. The used water containing various constituents such as dyes, chemicals is directly released into the sources of

water which gets contaminated and thus resulting into water pollution. Nowadays the use of synthetic fibres, polymers and finishes by textile industry is increasing at rapid rate. Since many of these products are resistant to biological degradation, it causes water pollution when released as effluent into the water sources.

Large amounts of highly coloured wastewater are discharged from textile, printing, paper, and leather industries which use many kinds of artificial composite dyes. These dye-containing industrial wastewaters discharged into streams and river constitutes one of the major sources of water pollution. Decolourising of textile and dye-manufacturing wastewater is currently a major problem for environmental managers. The coloured dye effluents are considered to be highly toxic to the aquatic biota and affect the symbiotic process by disturbing the natural equilibrium through reducing photosynthetic activity and primary production due to the colouration of the water in streams. Since most synthetic dyes have complex aromatic molecular structures which make them inert and biodegradable difficult when discharged into the environment. Coloured wastes are harmful to aquatic life in rivers, lakes and sea where they are discharged. Coloured water hinders light penetration and may disturb biological processes in water-bodies.

Moreover, dyes itself are highly toxic to some organisms and hence disturb the ecosystem. Dyes can cause allergic dermatitis, skin irritation, cancer, mutation, etc. In addition, biodegradation of some of them produce aromatic amines, which are highly carcinogenic. Treatment of the effluent from the dyeing and finishing processes in the textile industry is one of the most significant environmental problems. In practice, no single process provides adequate treatment and a combination of different processes is often used to achieve the desired water quality in the most economical way. Thus, there is a need to develop new decolourization methods that are inexpensive, effective and acceptable in industrial use.

2.2 Adsorption Process

Separation may be defined as a process that transforms a mixture of substances into two or more products that differ from each other in composition. The process is difficult to achieve because it is the opposite mixing, a process favoured by the second law of thermodynamics. Consequently, the separation steps often account for the major productions costs in chemical, petrochemical, and pharmaceutical industries. For many separation processes, the separation is caused by a mass separating agent. The mass separating agent for adsorption is adsorbent, or sorbent. Consequently, the performance of any adsorptive separation or purification process is directly determined by the quality of the sorbent (R.T. Yang, 2003).

Due to the progress made in sorbent and cyclic process developments, adsorption has already become a key separation tool that is used pervasively in industry. Adsorption is usually performed in columns packed with sorbent particles, or fixed bed adsorbers. The high separating power of chromatography that is achieved in a column is a unique advantage of adsorption as compared with other separation processes. The high separating power is caused by the continuous contact and equilibration between the fluid and sorbent phases. Under conditions free of diffusion limitation, each contact is equivalent to an equilibrium stage or theoretical plate. Usually several hundred to several thousand such equilibrium stages can be achieved within a short column. Thus, adsorption is ideally suited for purification applications as well as difficult separations. Partly because of this unique advantage, adsorption is well positioned to play a key role in a development of many future energy and environmental technologies (R.T. Yang, 2003).

Adsorption is the formation of a layer of gas, or solid, on the surface of a solid. The process of adsorption involves separation of a substance from liquid phase by accumulation or concentration onto the surface of solid phase. The adsorbing phase is the adsorbent, and the material concentrated or adsorbed at the surface of that phase is the adsorbate. Similar to surface tension, adsorption is a consequence of

surface energy. In a bulk material, all the bonding requirements (ionic, covalent, or metallic) of the constituent atoms of the material are filled by other atoms in the material. However, atoms on the surface of the adsorbent are not wholly surrounded by other adsorbent atoms and therefore can attract adsorbates.

Adsorption occurs in three steps. First step, the adsorbate diffuses from the major body of the stream to the external surface of the adsorbent particles. Second step, the adsorbate migrates from the relatively small area of the external surface to the pores within each adsorbent particle. The bulk of adsorption usually occurs in these pores because there is the majority of available surface area. Final step is when the contaminant molecule adheres to the surface in the pores. Adsorption at a surface is the surface which is the result of binding forces between the individual atoms, ions, or molecules of an adsorbate and the surface of the adsorbent. The adsorption process is generally classified as physisorption (characteristic of weak van der Waals forces) or chemisorption (characteristic of covalent bonding).

The differences of physisorption and chemisorption are molecules that are adsorbed by chemisorptions are very difficult to remove from the adsorbent. Whereas, the physically adsorbed molecules can usually be removed by either increasing the operating temperature or reducing the pressure. Chemisorptions also are a highly selective process. A molecule must be capable of forming a chemical bond with the adsorbent surface for chemisorptions to occur. Physical adsorption occurs under suitable conditions in most gas-solid system or in liquid-solid system.

Chemisorptions form only a monolayer of adsorbate molecules on the surface and stops when all reactive sites on the adsorbent surface are reacted. Physical adsorption can form multilayer of adsorbate molecules one atop another due to Van Der Waals forces. The chemisorptions rate increase with increasing temperature. The fundamental of adsorption is useful to distinguish between physical adsorption, involving only relatively weak intermolecular forces, and chemisorptions which